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THE ISOLATION OF AMERICIUM AND
CURIUM FROM IRRADIATED ^{241}AM TARGETS

by

W. MÜLLER, F. MAINO and J.-Cl. TOUSSAINT

1969



Joint Nuclear Research Center
Karlsruhe Establishment - Germany

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Luxembourg, April 1969 - 20 Pages - 5 Figures - FB 40

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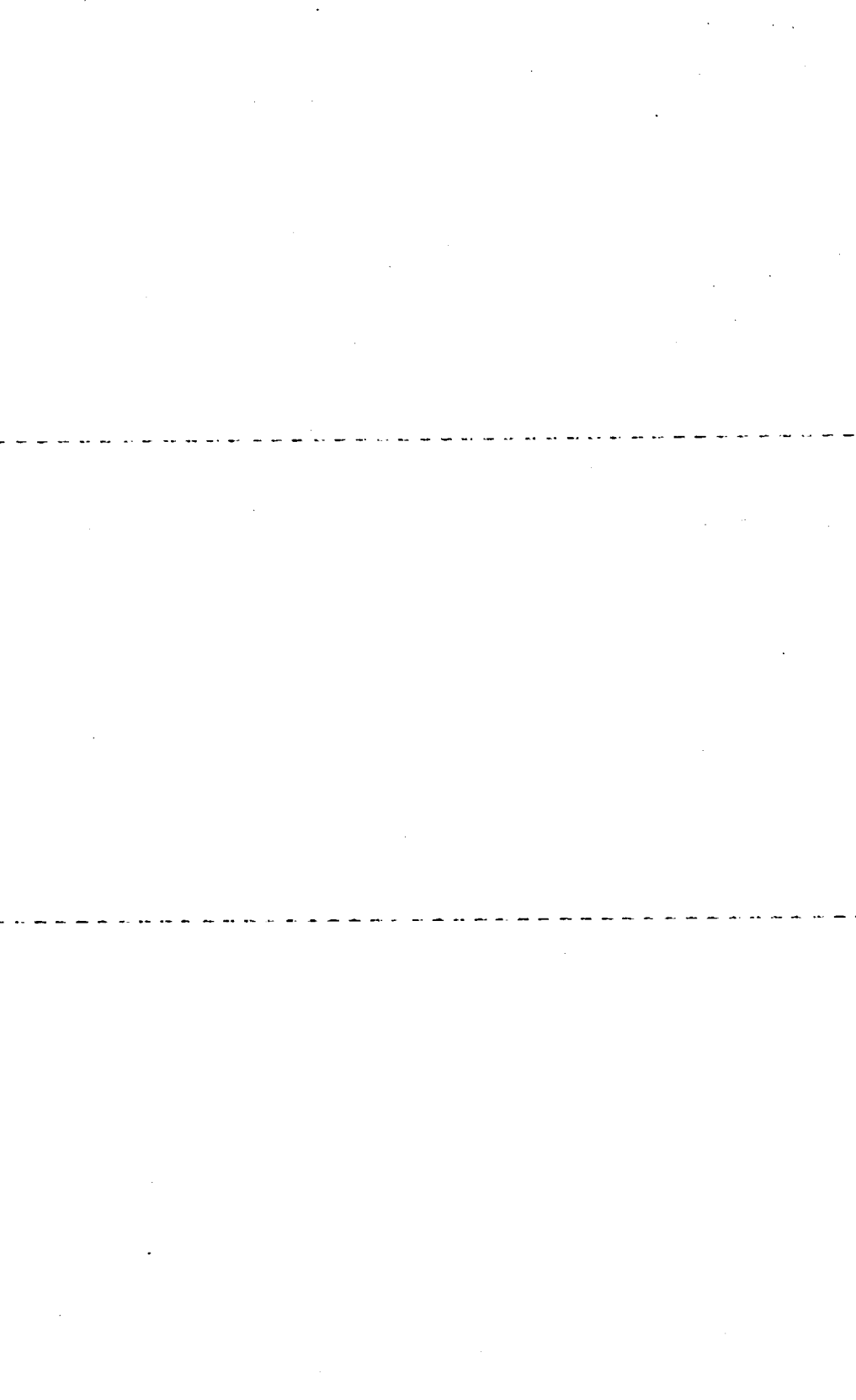
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ABSTRACT

At the European Transuranium Institute irradiated ^{241}Am targets were processed to isolate the heavy nuclides ^{238}Pu , ^{242}Pu , ^{241}Am and ^{242}Cm . The initial α -activity, mainly due to ^{242}Cm , exceeded 3 kCi.

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KEYWORDS

AMERICIUM 241	LITHIUM CHLORIDES
TARGETS	SOLUTIONS
IRRADIATION	PRECIPITATION
SEPARATION PROCESSES	OXALATES
PLUTONIUM 238	OXIDES
PLUTONIUM 242	PLATINUM
CURIUM 242	CAPSULES
ANIONS	WELDING
ION EXCHANGE	BATTERIES
HYDROCHLORIC ACID	RADIOISOTOPES

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1. INTRODUCTION (*)

A programme for the production of ^{244}Cm by irradiation of ^{241}Am in the reactor BR 2 was interrupted because one of the targets was damaged and ^{242}Cm was leaking into the cooling water of the reactor. In order to discover why this happened, metallographic investigations of several of the targets are being carried out at the present time. The pressure and composition of the gases formed during the irradiation are also being measured.

It seemed interesting to attempt to isolate the heavy elements from the target material not to be used for the above mentioned investigations. The purpose of this isolation was to find out if the equipment of the institute was suited or could be adapted to the handling of kCi amounts of α -emitters in concentrated form. In addition, it was expected to gain new information on shielding and containment. Such experience and knowledge are necessary to decide on a further programme for the production and isolation of ^{244}Cm and on the construction of shielded glove boxes for the handling of multigram quantities of americium and curium. Finally it was hoped to demonstrate the potential use of ^{242}Cm as a thermal power source.

2. THE IRRADIATION OF ^{241}Am

2.1. Irradiation targets

Each irradiation target contained initially 1.5 g ^{241}Am in the form of Al - AmO_2 (25 % by weight) pellets. The pellets had been pressed into aluminium claddings which were sealed by welding. Each capsule contained a gas plenum of about 1 cm^3).

2.2. Irradiation conditions

Seven targets arranged vertically in the reactor core of BR 2, and 2 targets positioned in the reflector were irradiated.

(*) Manuscript received on 17 December 1968.

ted at an average thermal flux of 2×10^{14} n/cm²sec. At the end of the irradiation, the integrated flux was approximately 10^{21} n/cm².

2.3. Irradiation products

After irradiation under the conditions mentioned in section 2.2., the actinide fraction still contained a part of the starting nuclide ²⁴¹Am, ²⁴²Pu, ²⁴²Cm and ²³⁸Pu as the decay product of ²⁴²Cm. It is impossible to calculate precisely the quantities of the heavy elements and fission products formed, as the effective flux, the flux distribution, the dependence of the capture or fission cross sections upon the neutron energy are only approximately known.

Table 1 shows how the amounts of heavy elements and fission products depend on the position of the targets in the reactor.

Table 1

	reactor core							reflector	
No. of the targets according to their position in the reactor	6	7	10	11	8*	12	1	13	15
Thermal power (W)	17	18.4	18.9	16.1	-	15.1	13.2	15.8	14.8
BY dose rate (10 ³ rem/h)	2	4	6	-	-	-	2	4	4

*) damaged target

Thermal power and BY dose rate of irradiated ²⁴¹Am targets

The thermal power and the dose rate of the irradiated targets were measured after a cooling period of about 250 days. The thermal power of each target decreased with a half-life of approximately 160 d, according to the α decay of ²⁴²Cm. The dose rate is determined by ²⁴¹Am, ²⁴²Cm and the fission products ¹³⁷Cs - ^{137m}Ba, ¹⁰⁶Ru - ¹⁰⁶Rh, ⁹⁵Zr - ⁹⁵Nb and ¹⁴⁴Ce - ¹⁴⁴Pr 2).

3. THE SEPARATION METHODS

3.1. Requirements

Before the processing, the α -activity of the ^{241}Am and ^{242}Cm present in the targets exceeded 3 kCi. These transplutonium elements were to be separated in a hot cell from ~ 150 g aluminium (matrix, cladding and part of the target holder), and several grams of plutonium (^{238}Pu , ^{242}Pu) and fission products (35.000 rem/h at contact).

A mixture of the americium and curium oxides was to be encapsulated and, after removal from the hot cell, inserted into a radioisotope power generator.

Because of the short half-life of the ^{242}Cm , a well proved processing method, which would not require further development and would permit a sufficient separation from the fission products and from the light elements, had to be chosen.

3.2. Separation scheme

The following scheme (fig. 1) is based on anion exchange^{3,4,5)}
^{6,7,8)} and has been improved on from experience gained during the processing of an americium target exposed to an integrated flux of 1.7×10^{22} n/cm² 9).

3.3. Problems

During the processing, problems which are characteristic for samples containing α -emitters of high specific activity like ^{242}Cm must be taken into account. Such problems are :

- α -radiolysis of aqueous solutions.

Hydrochloric acid solutions are decomposed by the α -radiation; H_2 , O_2 , Cl_2 are formed. Weakly acid salt solutions rapidly become acid deficient, and hydroxides are precipitated^{10,11)}.

Products of radiolysis oxidize cerium, and in neutral or basic solution even americium to the tetravalent state. Because aqueous solutions evolving radiolysis gases - 1 mg ^{242}Cm produces up to 60 ml (STP) of gas/day¹²⁾ - cannot be stored in

completely closed vessels, the danger of contamination is increased.

- Heat release of curium samples.

The α -decay heat of ^{242}Cm amounts to 120 W/g. In the bands of the exchanger, where curium concentrates, the resin and the aqueous solutions are decomposed by radiation and heat. Solutions and solid samples containing ^{242}Cm have to be cooled during storage. Despite external cooling, curium containing solids have been observed to become red hot by self-heating.

- Corrosion and erosion of material.

Among the metals, only tantalum withstands the attack of the highly corrosive solutions and atmospheres. High α -activity has been reported to cause sandlike silica to flake off from glass vessels¹¹⁾.

4. THE WORKING FACILITIES AND THE EQUIPMENT

4.1. Process cell

All handling and processing of the irradiated targets were to be carried out in only one shielded cell. A steel caisson, a prototype of the high activity cells described previously¹³⁾, was fitted out for the chemical separations, for the preparation of samples for metallographic investigations, and for the encapsulating of the oxide mixture. Therefore, a water supply with heat exchanger for a closed cooling system, inlet valves for gases, special sockets for a hack-saw, a polishing machine, an ultrasonic cleaner, a welding machine etc. were provided.

4.2. Equipment

For equipment such as the dissolving vessel, evaporator, gas absorption system, pipetter, stand for heatable exchanger column, manometer, almost exclusively glass, polyvinylchloride or plexiglass were used. The exchanger column was improved : the resin bed (Dowex 1 x 8, 100 - 200 mesh, length and dia-

meter 30 and 4 cm respectively) was compressed by porous polyethylene discs which prevented the formation of large gas bubbles during the separations¹⁴⁾.

4.3. Radioactivity measurement and monitoring

In order to measure the radioactivity inside the cell,

- an ionization chamber,
- a BF_3 neutron detector, and
- solid state surface barrier detectors¹⁵⁾ were used.

The BF_3 counter permitted the measurement of the neutrons emitted by curium. A solid state detector was placed at the exit of the exchanger column and served to monitor continuously the separations.

The process cell was connected to the general nitrogen ventilation circuit of the hot cell wing. This required a continuous measurement of the radioactivity behind the absolute filters and the charcoal bed to detect any airborne activity which might contaminate the other metallographic cells connected to the same circuit. In the operating area, the air near the manipulator ports of the process cell was sucked through paper filters which were continuously monitored.

5. THE ISOLATION AND PURIFICATION OF THE AMERICIUM-CURIUM FRACTION

5.1. Preliminary experiment

Equipment, material and separation methods were checked by processing one target.

Fig. 2 indicates the separation of plutonium and the fission products $^{106}\text{Ru} - ^{106}\text{Rh}$ and $^{95}\text{Zr} - ^{95}\text{Nb}$ from the trivalent actinides and lanthanides by sorption on Dowex 1 x 8 exchanger. The plutonium fraction is stripped with 0.5 M HCl. The curves show the variation of the total α and γ activities of the different fractions during the column run.

Fig. 3 shows, again as a variation of the activities, the separation of the lanthanides by sorption of the actinides on Dowex 1 x 8 from 10 M LiCl, 0.1 M HCl, 0.1 M hydroxylammonium chloride, 5 % by volume of CH₃OH. Americium and curium are stripped with 8 M HCl.

This americium - curium fraction, isolated from the first target, was precipitated as oxalate and served to carry out shielding measurements necessary to calculate the dimensions of the neutron and gamma shielding of the heat source. Table 2 summarizes some of the results obtained with a mixture of about 0.1 g ²⁴²Cm and 1.0 g ²⁴¹Am at a distance of 57 cm between the source and the detector.

Table 2

	Thickness of water layer (cm)				
	5	10	15	20	25
neutron dose rate (mrem/h)	10	3.8	1.3	0.7	0.4
BY dose rate (mrem/h)	100	50	35	25	20

Neutron and BY dose rate of a mixture of americium and curium shielded by water (from measurements by R.Puel)

5.2. Main separation

The bulk of the irradiated material was processed according to the results and the experience gained from the first target. After dissolving the targets, the plutonium (approximately 6 g with a ²³⁸Pu/²⁴²Pu mass ratio of 4) was eliminated on **one** single column. The separation of the lanthanides from the trivalent actinides was carried out with half of the material per column.

The purification which was achieved under these conditions can be estimated from fig. 4, The upper curve represents a part of the Y spectrum of the hydrochloric acid solution before

the separation of the plutonium; the lower curve shows the corresponding part of the spectrum after the separation of the lanthanides. The decontamination factors of the predominating fission products are of the order of 10^3 .

5.3. Comments

5.3.1. Dissolution

In order to prevent the formation of detonating gas, it has often been recommended to dissolve aluminium cladding and matrix using a mixture of sodium hydroxide and nitrate. However, as the process cell was flushed with nitrogen, the targets could be dissolved in pure sodium hydroxide solution. The residue obtained was readily soluble in concentrated hydrochloric acid. It is important to eliminate the aluminium as completely as possible, since otherwise a precipitate may form in concentrated hydrochloric acid or lithium chloride solution.

5.3.2. Plutonium separation

Potassium nitrite is added before the plutonium separation. If excess nitrite is destroyed by amidosulfonic acid, the gas formation on the exchanger column appreciably decreases.

5.3.3. Separation of lanthanides and actinides

It is general practice, to load the mixture of the trivalent actinides and lanthanides from at least 12 M LiCl solution to obtain narrow sorption bands and, hence, achieve a good group separation during the elution with 10 M LiCl solution. Because of the high α -activity, however, LiCl solutions of the same composition were used for both loading and elution :

10 M LiCl, 0.1 M HCl, 0.1 M hydroxylammonium chloride, 5 % by volume CH_3OH . With the addition of methanol, the acid depletion is slowed down, and the loading concentration of curium can be increased to 250 W/l. The broadening of the americium - curium band on the exchanger column reduces the radiation damage of the resin and lowers the gas evolution in the resin bed. The reductant suppresses the oxidation of cerium.

5.3.4. Lithium removal

The stripping of the americium - curium fraction with 3 M HCl results in a solution which contains up to 2 M LiCl. After precipitation of the actinide hydroxides the residual lithium content can easily be removed by washing.

5.3.5. Oxalate precipitation

The purified americium - curium fraction was precipitated from 0.1 M HNO₃ with oxalic acid. Immediately after filtering and drying, the precipitate was placed in a platinum crucible. Despite external cooling of the crucible, the mixture of the oxalates decomposed into oxides and became red hot.

A total of 4.3 g of the oxides containing approximately 400 mg of ²⁴²Cm were loaded into a double walled platinum capsule which was sealed by welding¹⁶⁾.

6. CONCLUSIONS

The experience obtained during the processing work described above may be summarized as follows :

6.1. Hot cells

The high activity cell permitted the safe handling of kCi amounts of α -emitters. Caisson, joints and filters corresponded to the requirements. The manipulator sleeve is the most vulnerable part of the equipment. It should be protected against mechanical damage from the manipulator wrists. The latex parts of the sleeves deteriorate because of attack by ozone. Despite severe damaging during a shutdown of the nitrogen ventilation system resulting in ruptures of the latex (fig. 5), the contamination was limited to the surface of the manipulator arm.

Because of Cm aerosols, the contamination inside the process cell increased to such a value that microsampling for ana-

lysis became impossible. In future, the use of a second cell for analysis and further processing of fractions already purified will be necessary.

6.2. Material

The behaviour of the glass, polyvinyl chloride or plexiglass equipment was satisfactory. In contact with highly active solutions, rubber and PVC tubing were rapidly destroyed, whereas silicon rubber resisted the attack for a longer time. Only slight "α-erosion"¹¹⁾ of the glass vessels could be detected.

6.3. Methods

Anion exchange as described in section 5.2. is a useful method for the isolation of kCi's of transplutonium elements from irradiated targets.

The separation of Pu (Zr, Ru) can be simplified

- either by batch extraction with solutions of substituted ammonium salts¹⁷⁾
- or by ion exchange using a slurry technique.

As starting material for the preparation of oxide powders, oxalates can be precipitated from highly active curium or americium-curium solutions.

6.4. Shielding

Up to gram amounts of ^{242}Cm can be safely handled behind a water shield of 25 cm.

Acknowledgements

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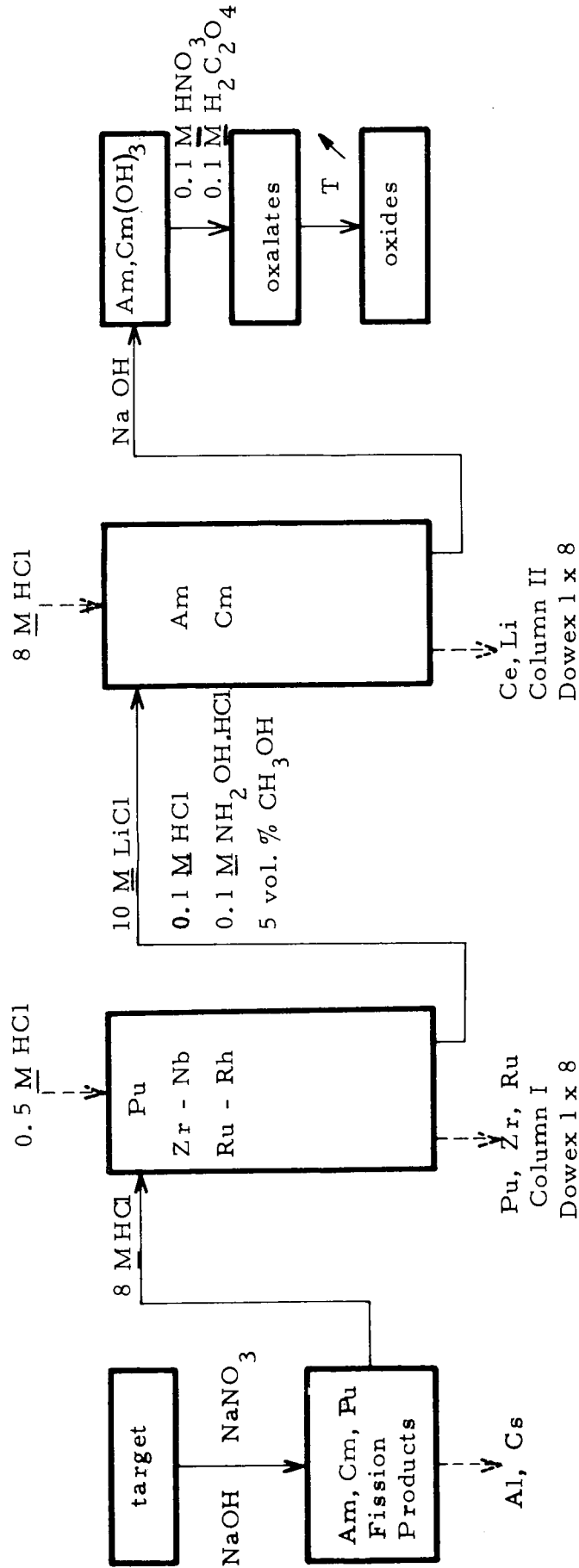


Fig. 1 Flow-Sheet for the isolation of transplutonium elements from irradiated ^{241}Am targets

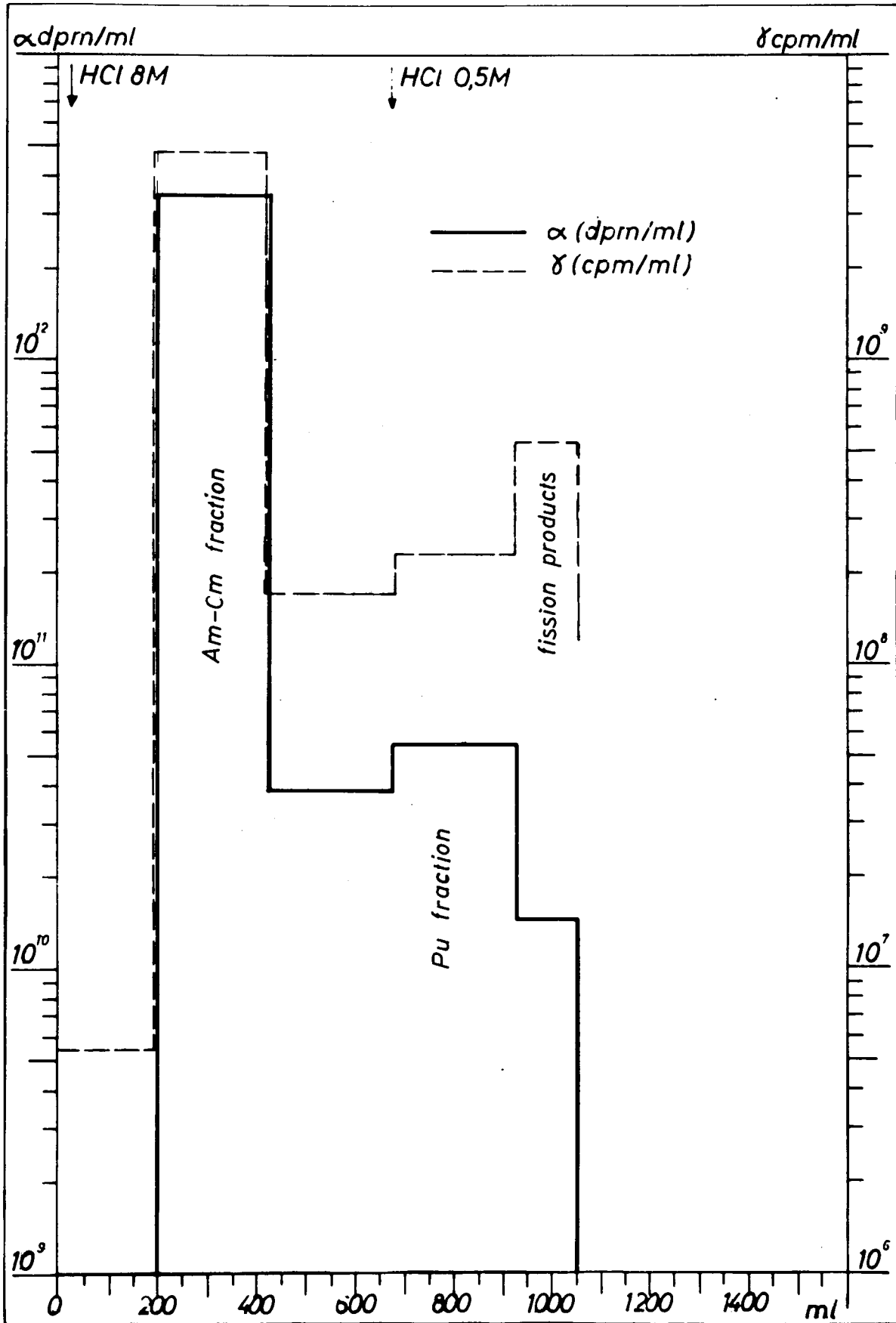


Fig. 2 Separation of plutonium and fission products (Zr-Nb, Ru-Rh) from americium, curium and lanthanides on Dowex 1 x 8.

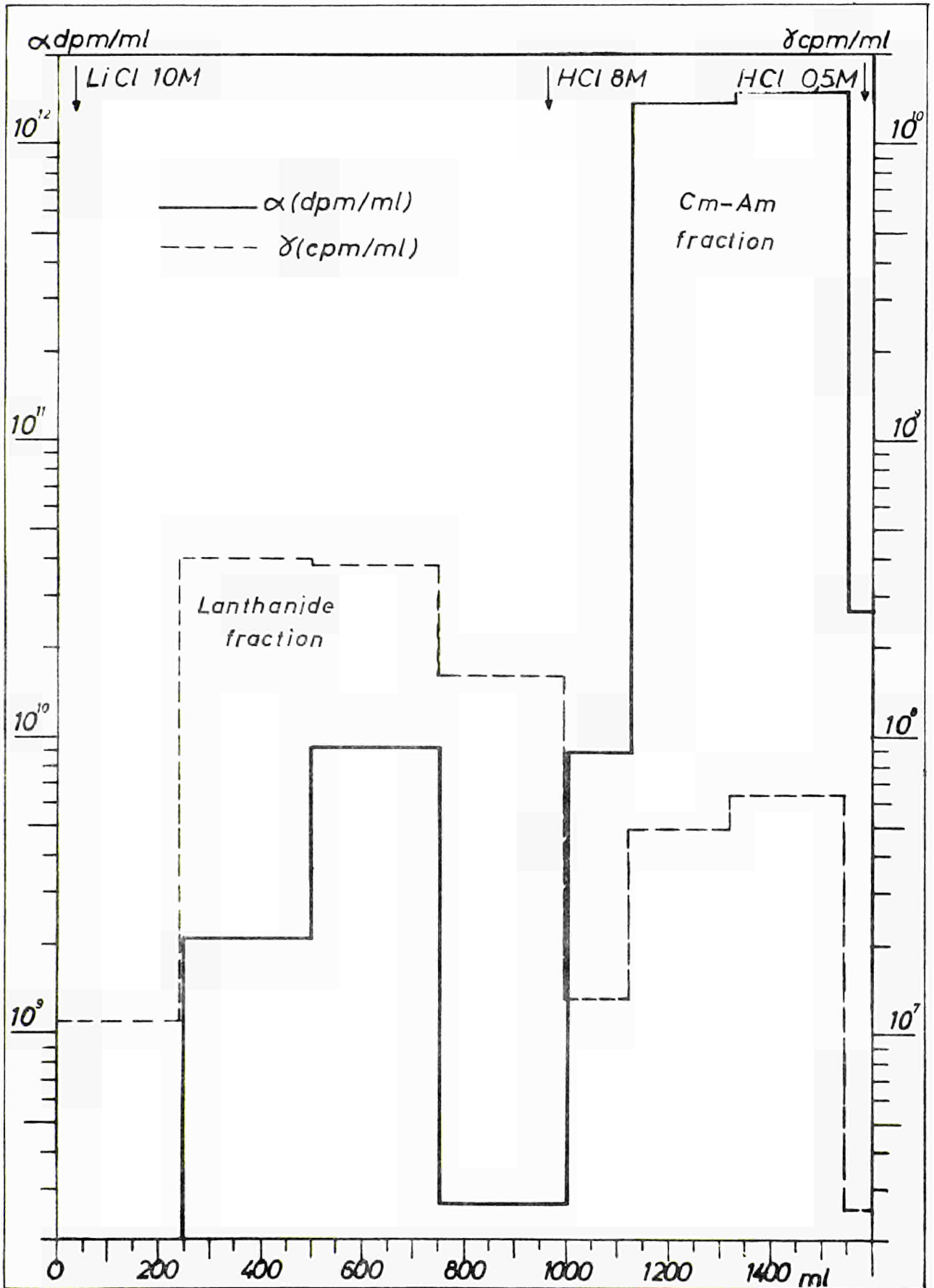


Fig. 3 Separation of lanthanide fission products (Ce-Pr) from the americium-curium fraction on Dowex 1 x 8.

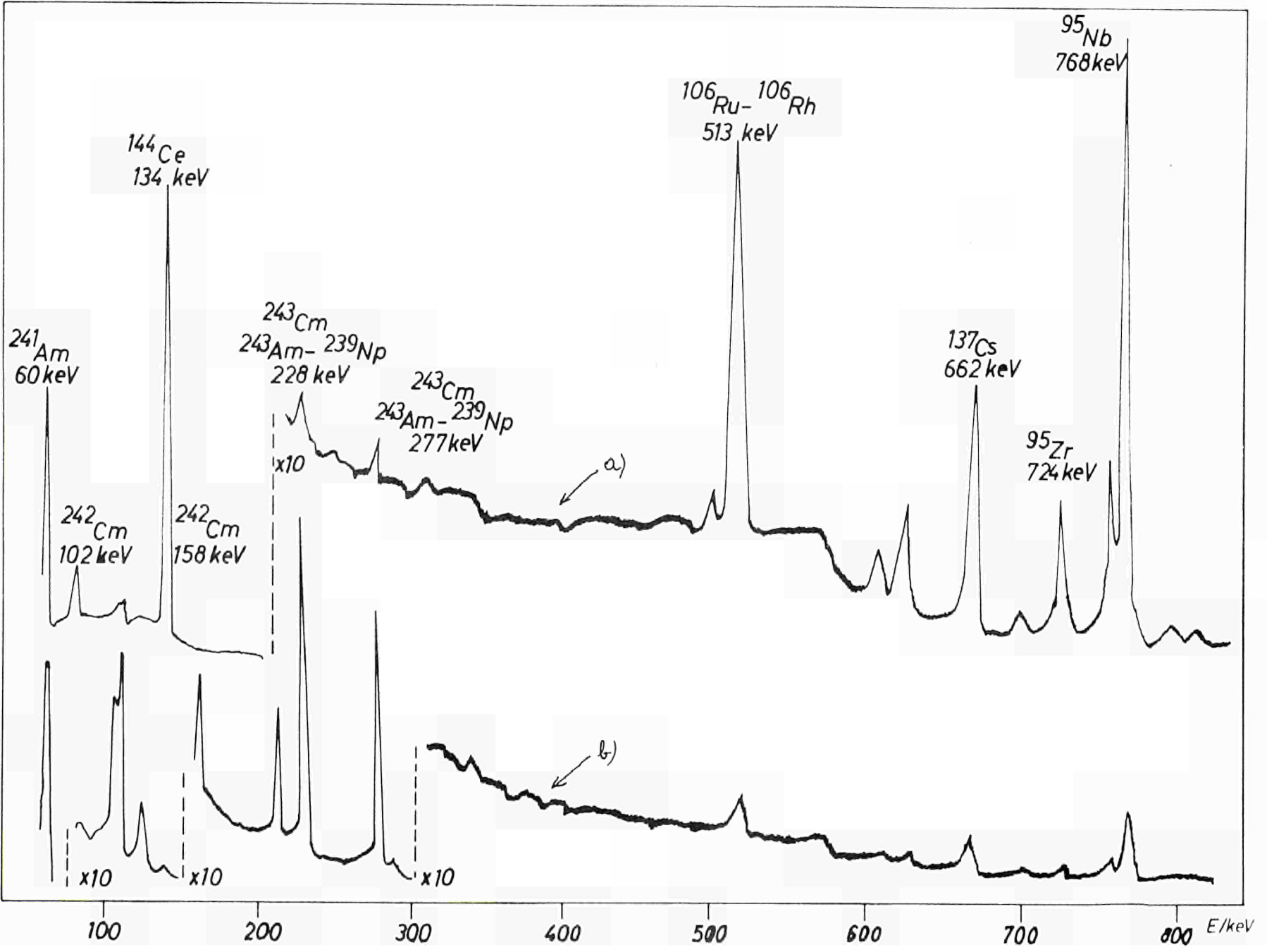


Fig. 4
 Gamma spectra of the hydrochloric acid feed solution before plutonium removal (a) and of the americium-curium fraction after lanthanide separation (b).

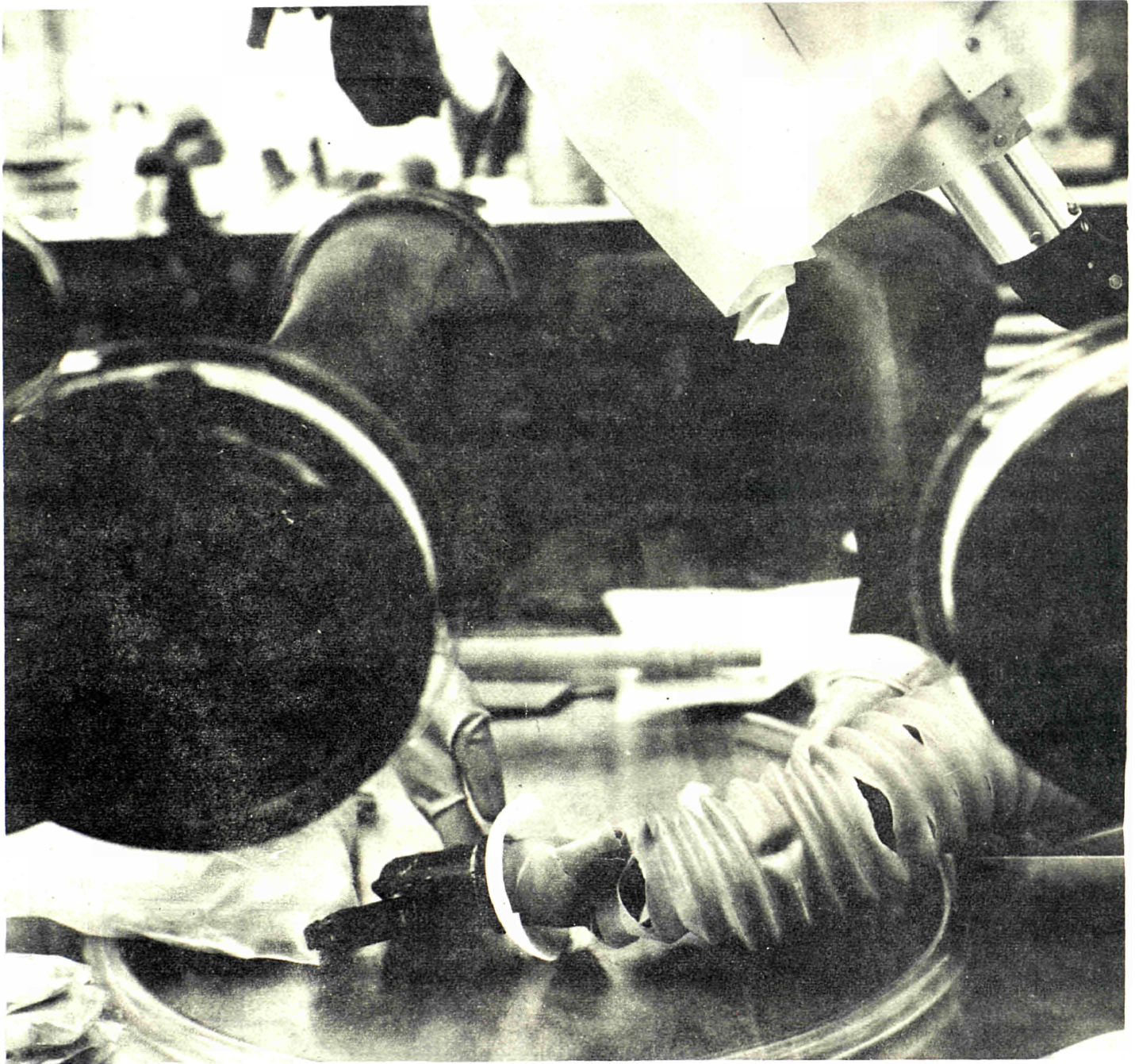


Fig. 5

Ozone attack on manipulator sleeve

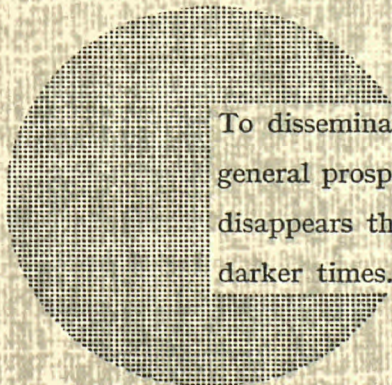
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Alfred Nobel

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